BETA-SPECTRA OF IODINE

By

GERALD F. TAPE

A THESIS

SUBMITTED IN PARTIAL FULFILLMENT OF THE REQUIREMENTS
FOR THE DEGREE OF DOCTOR OF PHILOSOPHY
AT THE UNIVERSITY OF MICHIGAN

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Beta-Spectra of Iodine

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Artificial activation of tellurium and iodine by deuterons and neutrons yields four radioactive isotopes of iodine. The momentum distributions of the beta-particles emitted by these isotopes were examined in a large hydrogen-filled cloud chamber placed in a uniform magnetic field. Applications of the Fermi theory of beta-decay and of the Konopinski-Uhlenbeck modification were made and the extrapolated end points were compared to the inspection end points. Gamma-radiation was found with each activity. An estimate of energies of the gamma-radiation associated with I^{130} and I^{131} was determined from the electrons ejected from a lead absorber placed in the cloud chamber. The following results were obtained:

Half-life	Probable Isotope	Energy of Radiations: Beta	Energy of Radiations: Gamma
25 ±1 minutes	I^{128}	2.40 ±0.07 Mev	(Yes)
13.0±0.3 days	I^{126}	1.20 ±0.03	(Yes)
8.2±0.3 days	I^{131}	0.687±0.010	0.27 Mev 0.46
12.5±0.5 hours	I^{130}	0.83 ±0.03	0.59

The original Fermi theory is in better agreement with the experimental data, the Kurie plots being linear over a considerable range to the end-point.

INTRODUCTION

THE original work of Fermi and his associates[1] showed that tellurium and iodine were made radioactive when irradiated with slow neutrons. An activity of 60 minutes half-life was identified in tellurium when it was irradiated with 17-Mev gamma-rays[2] and with lithium neutrons.[3] Pool, Cork and Thornton[4] found an additional activity produced by fast neutrons in tellurium of about 30 days. Chemical identification of these two activities with tellurium and the discovery of activities of 10 hours and 8 days half-lives produced by deuteron bombardment of tellurium were included in a report by Tape and Cork.[5] The formation of an activity of 13 days half-life produced by fast neutron bombardment of iodine was also reported. Additional work on the isotopes of iodine was done by Livingood and Seaborg[6] producing iodine activities having half-lives of 4.0 days, 13 hours, and 8 days in addition to the 25-minute and 13-

* Now at the Department of Physics, Cornell University.
[1] Amaldi, D'Agostino, Fermi, Pontecorvo, Rasetti, Segrè, Proc. Roy. Soc. **A149**, 522 (1935).
[2] W. Bothe and W. Gentner, Zeits. f. Physik **106**, 236 (1938).
[3] F. A. Heyn, Nature **139**, 842 (1937).
[4] Pool, Cork and Thornton, Phys. Rev. **52**, 239 (1937).
[5] G. F. Tape and J. M. Cork, Phys. Rev. **53**, 676 (1938).
[6] J. J. Livingood and G. T. Seaborg, Phys. Rev. **54**, 775 (1938).

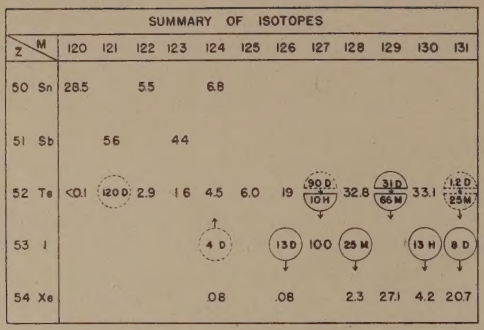

FIG. 1. Summary of stable and radioactive isotopes of iodine and tellurium.

day activities. Seaborg, Livingood and Kennedy[7] have shown that there are three pairs of radioactive tellurium isomers. The stable and radioactive isotopes of tellurium and iodine are shown in Fig. 1. Activities enclosed in solid lines have been verified in this laboratory.

The three tellurium activities of 66 minutes, 10.0 hours, and 31.5 days were produced by bombardment of tellurium with deuterons of energies up to 9 Mev and with fast neutrons. The 10-hour activity was also produced by fast neutron bombardment of iodine. The assignment of this activity to Te^{127} is made through $I^{127}(\eta, H^1)Te^{127}$. The assignment of the two other activities is made to Te^{129} since they are not formed by fast neutron bombardment of iodine but are formed by similar bombardment of tellurium or by irradiation with gamma-rays.[2] All radioactive isotopes of iodine and tellurium except Te^{121} and I^{124} have been definitely shown to emit negative beta-particles.

The beta-ray spectra of all iodine activities produced by deuteron and neutron activation of tellurium and iodine were measured. The momenta of the beta-particles emitted during decay of the iodine isotopes were measured in a twelve-inch cloud chamber placed in a magnetic field constant to 0.5 percent over the volume of the chamber. The control circuit of the vacuum tube type was designed by Richardson.[8] The chamber filled with hydrogen to a pressure slightly in excess of atmospheric was photographed by a single camera. The tracks thus photographed were reprojected to their original size and measured. Restrictions were placed on track selection to insure no scattering of the beta-particle, its origin in the radioactive sample, and a path perpendicular to the magnetic field and to insure against subjective weighting of certain energy groups.

All radioactive samples were prepared by deuteron or neutron bombardment in the University of Michigan cyclotron. After the samples were bombarded and the appropriate chemical separations had been performed, the activated material was finely divided and spread on filter paper. The sample was coated with a solution of paralodion dissolved in alcohol and ether and, after drying, was cut to a length of 3 cm and to a width determined by its activity, the width in all cases being less than 0.5 cm. The mounting frame inside the chamber was made from copper strip and cemented to the wall of the chamber. The frame was made in the shape of a C with the sample placed across the open end and the body of the C next to the chamber wall. By mounting the sample on the arms of the C-frame, there was only filter paper immediately behind the active material, and the glass wall of the chamber was at a minimum of 2 cm behind the paper. Back scattering was thus materially reduced. The filter paper mounting weighed 7.5 mg/cm².

IODINE[128]

This well-known slow neutron period having a half-life of 25±1 minutes was first observed by Fermi and his co-workers.[1] Since iodine has but a single stable isotope, I^{127}, this activity has been

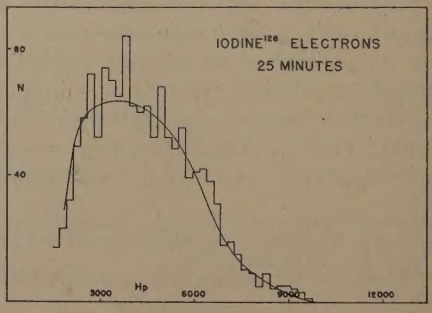

FIG. 2. Momentum distribution of electrons from I^{128}.

[7] Seaborg, Livingood and Kennedy, Phys. Rev. **55**, 794 (1939).

[8] J. R. Richardson, Rev. Sci. Inst. **9**, 152 (1938).

definitely assigned to I^{128} through neutron capture. Livingood and Seaborg[6] produced the same radioactive isotope by bombarding tellurium with 8-Mev deuterons. This reaction has been checked in this laboratory with 9-Mev deuterons.

Since Te^{127} is unstable, the transmutation must be $Te^{126}(H^2, \gamma)I^{128}$ or $Te^{128}(H^2, 2n)I^{128}$ with the latter reaction more likely. I^{128} decays to Xe^{128} with the emission of negative beta-particles and gamma-rays.

Absorption measurements in aluminum[6] indicated a maximum energy of 2.2 Mev. Using a magnetic spectrometer, Alichanian, Alichanow and Dzelepow[9] determined the upper end-point of the I^{128} spectrum to be at 2.1 Mev. Bacon, Grisewood and van der Merwe[10] using a cloud chamber found a complex spectrum. Their analysis of a K-U application showed end-points at 1.2 and 2.1 Mev.

For cloud-chamber work samples of C.P. lead iodide were bombarded for 10 microampere minutes with 7-Mev deuterons. These samples were mounted in the chamber without performing any chemistry. The decay of a fraction of the activated sample was followed at the same time that photographs were being taken. The decay curve showed a pure period of 25 minutes during the period of photography and no contamination due to the presence of the lead. Since the half-life of I^{128} is only 25 minutes, several samples had to be used to obtain a large number of tracks. Of all samples used to obtain the momentum distribution of the beta-particles, the maximum surface density used was 15 mg/cm². This does not include the paper mounting of 7.5 mg/cm².

The momentum distribution of the 1330 measured tracks is shown in Fig. 2. The magnetic field was maintained at 448 oersteds. The inspection end-point is at 9680 $H\rho$ or 2.44 Mev. Below 2500 $H\rho$ the spectrum has very little significance. That this is true comes from the criteria used in track selection. From the demand that a track be at least 10 cm long to minimize the error due to the use of a single camera, it is seen that for a field of 448 oersteds the lower end of the spectrum loses significance around 2500 $H\rho$.

Taking the expressions developed by Fermi and by Konopinski and Uhlenbeck for the probability of emission of a beta-particle having a momentum between η and $\eta+d\eta$, Kurie, Richardson and Paxton[11] have shown that the expression $(N/f)^{1/k}$ plotted against the kinetic energy of the beta-particle will give a straight line if the theory be valid. N is the number of particles in a given momentum interval, f is a function of Z and η, and k equals 2 for the Fermi theory and 4 for the

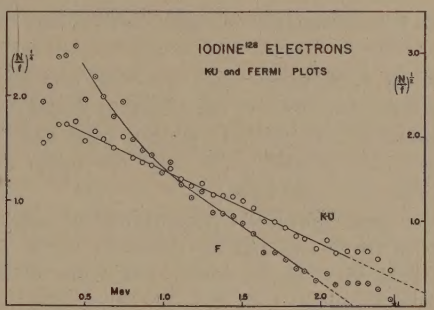

FIG. 3. Fermi and K-U treatments of I^{128} electrons.

Konopinski-Uhlenbeck modification. An approximation given by Bethe and Bacher[12] was used to evaluate f for iodine, where Z equals 53. This evaluation takes care of the Coulomb effect of the nucleus on the emitted particle.

The two plots showing the application of the Fermi and Konopinski-Uhlenbeck theories to the beta-spectrum of I^{128} are shown in Fig. 3. The points shown on K-U and Fermi diagrams are points determined from actual experimental data and not from the distribution curves, which are averages of the data. Thus the same statistical fluctuations appear in the K-U and Fermi curves as appear in the momentum histograms. It was hoped that the use of such a method would give an additional means of averaging the collected data and yield an independently determined end-point. If the uncertainty in each point is taken into account, one sees that for momentum intervals in which only one track is measured, the ordinate of the histogram would have a spread in value between 0 and 2 with a most probable

[9] Alichanian, Alichanow and Dzelepow, Physik. Zeits. Sowjetunion **10**, 78 (1936).
[10] Bacon, Grisewood and van der Merwe, Phys. Rev. **54**, 315 (1938).
[11] Kurie, Richardson and Paxton, Phys. Rev. **49**, 368 (1936).
[12] H. A. Bethe and R. F. Bacher, Rev. Mod. Phys. **8**, 194 (1936).

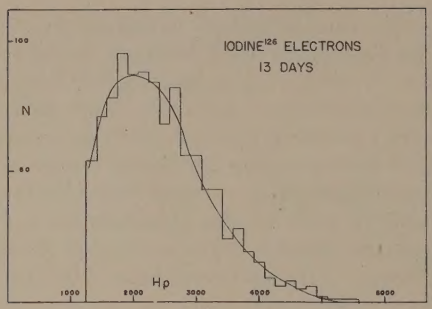

FIG. 4. Momentum distribution of electrons from I^{126}.

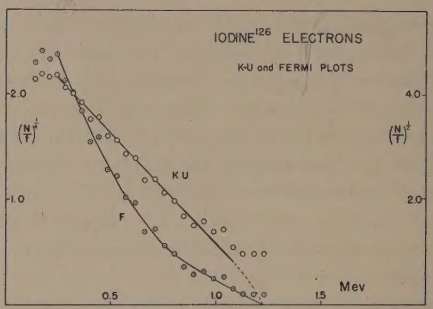

FIG. 5. Fermi and K-U treatments of I^{126} electrons.

value of 1. Similarly with the K-U and Fermi treatments, for $N=1$, the ordinate would have a spread of $2(1/f)^{1/k}$. It is thus seen that the uncertainty of each point near the upper end-point is extremely large.

The upper end-points determined by applications of the Fermi and K-U theories are at 2.20 and 2.94 Mev, respectively. The value of 2.94 Mev, determined by extrapolation, is definitely too high, in fact more than 20 percent higher than the maximum energy measured. The value of 2.20 Mev for the Fermi end-point was also determined by a short extrapolation. Since there were energies measured greater than 2.2 Mev, one must assume that either 2.2 Mev is too small and does not represent the true end-point of I^{128} or that it is all right and that the higher energy tracks are due to some contamination. The only contamination that might be present would be Al^{28} from the foil covering the PbI sample during bombardment. Its half-life is 2.3 minutes, and it emits negative betas with energies up to 3.3 Mev and gamma-rays having an energy of about 2.3 Mev. However, the decay curve did not show the presence of any short life activity. If radioactive aluminum were present, one would expect to measure energies up to 3.3 Mev. Since the upper limit of the spectrum as measured ends at 2.44 Mev, it would seem that the absence of higher energy particles shows the absence of Al^{28} contamination.

IODINE^{126}

When iodine is bombarded with fast neutrons ($Li+H^2$), an iodine activity of 13.0 ± 0.3 days half-life is formed.[5] This activity chemically identified as iodine and checked by alpha-particle bombardment of antimony[6] is undoubtedly I^{126} formed by $I^{127}(n, 2n)I^{126}$. It is negative active and decays to stable Xe^{126}. The upper end-point of the beta-ray spectrum has been placed at 1.13 Mev by Livingood and Seaborg[6] who made absorption measurements in aluminum.

$NaIO_3$ was bombarded in the cyclotron with neutrons from the $Li+H^2$ reaction. The activated iodine was separated from the bulk of the $NaIO_3$ by the Amaldi modification[1] of the Szilard-Chalmers reaction. The concentrated sample prepared for cloud-chamber mounting as previously described had a surface density of 22 mg/cm^2. The sample was allowed to age for two days so that the entire activity would be of 13 days half-life.

The momentum distribution of the 1060 measured tracks is shown in Fig. 4. A field strength of 336 oersteds was used giving a maximum radius of curvature of 16 cm. By inspection the upper end-point of the I^{126} beta-ray spectrum is at 4300 ± 100 $H\rho$. This corresponds to a maximum energy of 1.20 ± 0.03 Mev. N, the number of tracks, is insignificant for values of $H\rho$ less than 1260. The shape of the spectrum is that of the general single beta-ray spectrum. Applications of the Fermi and K-U theories were made and the Kurie plots are shown in Fig. 5. The K-U end-point obtained by extrapolation from the region between 0.3 and 0.9 Mev is at 1.29 Mev, a difference of 7.5 percent as compared with the inspection upper limit of 1.20. The Fermi curve is concave upward over its major portion and from 0.9 Mev to the end-point it is best represented by a straight line. The Fermi end-point thus determined is at 1.22 Mev, 1.6 percent

higher than the end-point determined from the momentum histogram.

There are only two momentum intervals in which N differs markedly from the distribution curve (Fig. 4). These occur at 2520 and 2688 $H\rho$. Since these two variations are in opposite senses, it seems logical that a preference for one interval existed over the other. 2688 $H\rho$ corresponds to an energy of 0.44 Mev. If this point in the distribution corresponds to the conversion electrons from the K shell of Xe, the internally converted gamma-ray would have an energy of 0.48 Mev. This agrees with the value of 0.5 given by absorption in Pb.[6] However, such conclusions as to the identification of the electrons in the interval at 2688 $H\rho$ with conversion electrons seems impossible in the light of statistical fluctuations.

Iodine^{131}

The 8-day activity produced when tellurium is bombarded with deuterons and originally associated with Te^{131} has since been assigned to I^{131}. This activity is produced directly by deuteron bombardment of tellurium and also by decay of Te^{131} to I^{131} with the emission of a negative beta-particle. That it is formed directly is verified by the intensity ratios of the initial iodine separation to subsequent separations from the active tellurium. The half-life has been measured at 8.2±0.3 days. I^{131} decays with the emission of negative beta-particles to Xe^{131}. Gamma-rays have also been detected.

For cloud-chamber work, powdered tellurium metal prepared by pulverizing stick tellurium was bombarded in a vacuum with 10 microampere hours of 9-Mev deuterons. After bombardment the tellurium was placed in a distilling flask and a trace of KI added as a carrier for the active iodine formed during bombardment. The iodine was separated by oxidation and distilled off. The separated iodine was precipitated as AgI. The small amount of precipitate was evenly distributed over the surface of the filter paper. The solution containing the tellurium was neutralized with NH_4OH and the Te was precipitated by reduction to the free state with $SnCl_2$.

Two samples of AgI obtained from two bombardments of tellurium were used to determine the beta-spectrum of I^{131}. From the first sample weighing 10 mg/cm^2, over one thousand tracks were examined and an end-point of 0.692±0.015 Mev was obtained. More recent work with a sample having a surface density of 2 mg/cm^2 showed no essential differences. Tyler and Lawson[13] found that the experimentally determined shape of a beta-ray spectrum was a function of the thickness of the source. The two independently obtained spectra were normalized to the same number of tracks and the shapes of the two were found to be the same within the limit of statistical fluctuations.

As a result of the comparison of the data from the two samples, the data were combined and the resulting spectrum of I^{131} is shown in Fig. 6. Over 2000 tracks are represented in the histogram. The inspection end-point for this resultant histogram is at 0.687±0.010 Mev. The K-U and Fermi applications are shown in Fig. 7. The Fermi limit is within the range of the inspection end-point while the K-U extrapolation occurs at

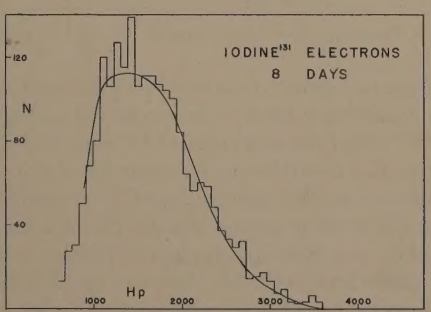

Fig. 6. Momentum distribution of electrons from I^{131}.

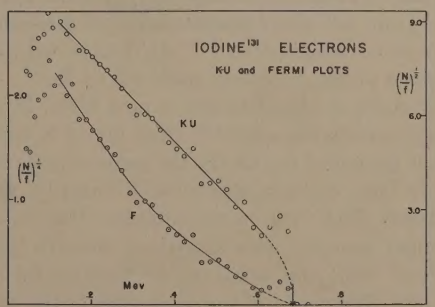

Fig. 7. Fermi and K-U treatments of I^{131} electrons.

[13] A. W. Tyler, Phys. Rev. **56**, 125 (1939); J. L. Lawson, Phys. Rev. **56**, 131 (1939).

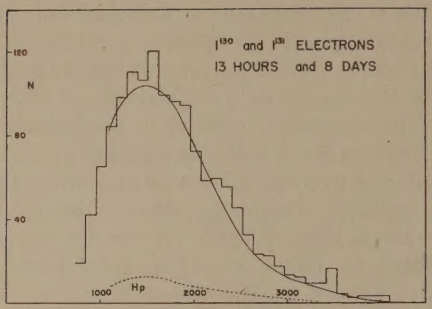

FIG. 8. Solid curve—momentum distribution of electrons from I^{130}. Broken curve—momentum distribution of electrons from I^{131}.

0.78 Mev, about 11.5 percent high. The curve shapes are the same as have been found for single spectra. With the curves as drawn, there are too many tracks in the vicinity of the end-point. It may be that there are too few tracks in the region of 0.6 Mev and thus the upper limit should be extended to slightly higher values, perhaps as high as 0.72 Mev. From examination of the rest of the curve this does not seem very likely. The value of 0.69 Mev does not compare favorably with 1.24 Mev found from absorption measurements.[6]

IODINE^{130}

The formation of an iodine isotope of about 13 hours half-life from tellurium bombarded with deuterons was discovered by Livingood and Seaborg.[6] This activity has been checked and the half-life determined to be 12.5±0.5 hours. Since this activity was not found in neutron activated samples of iodine or in subsequent iodine separations from tellurium bombarded with deuterons, it must be assigned to I^{130}. It decays with the emission of negative beta-particles to Xe^{130}.

I^{130} could not be obtained in pure form. It was always associated with I^{131} (half-life of 8 days). It was prepared in exactly the same way as was I^{131}. In fact, the beta-spectra of I^{130} and I^{131} were obtained from the same sample. The cloud-chamber sample had a surface density of 2 mg/cm^2 with the usual paper backing of 7.5 mg/cm^2. The decay of the remainder of the total sample was followed with an ionization chamber and electrometer. From the decay curves it was found that at the time the I^{130} spectrum was measured, the ratio of activities (I^{130} : I^{131}) was 10 to 1.

The beta-ray histogram of the combined spectra is shown in Fig. 8. From the histogram alone it would be difficult to tell that two activities are represented. The difference between this histogram and that of I^{131} (Fig. 6) is in the upper end-point. The maximum energy measured was 0.69 Mev for I^{131} and is here 0.83 for the combined spectra. These two values were obtained for the same sample of active iodine. A period of one week separated the measurement of the two spectra. Thus it must be assumed that the two spectra are almost identical with I^{130} having an upper end-point at 0.83±0.03 Mev.

That the histogram in Fig. 8 is composite is shown by the K-U treatment (Fig. 9). It has been seen that K-U curves have been best represented by straight lines over their major portions. Here the K-U curve is definitely concave upward showing the presence of more than one component. The Fermi curve is also more concave than for a single spectrum. Two straight lines have been drawn through the high and low energy regions of the K-U curve to show its complexity. The extrapolations of these two lines do not

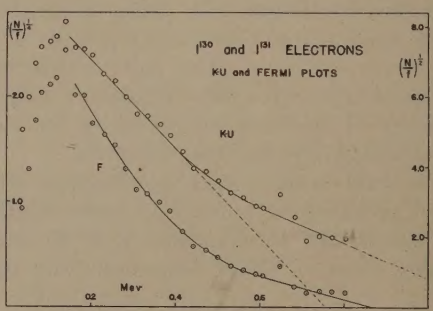

FIG. 9. Fermi and K-U treatments of I^{130} and I^{131} electrons.

indicate end-points. One concludes definitely that two spectra are present and from the histogram in Fig. 8 that the end-point of I^{130} is at 0.83 Mev. In Fig. 8, a contribution of about 0.1 of the total number of tracks (broken curve) was subtracted from the total to obtain the distribution for I^{130} (solid curve). Absorption measurements[6] showed an upper limit of 1.05 Mev for the I^{130} beta-ray spectrum. This is considerably larger than 0.83 Mev. Both methods of measurement indi-

cated that the upper limit of I^{130} and I^{131} are at about the same energy. However, the two methods differ in results in that they yield different values for the upper end-points and opposite signs for the energy difference between the two. The cloud-chamber work places I^{130} with a somewhat higher maximum energy.

Gamma-Rays

An attempt was made to measure the energies of the gamma-rays associated with each isotope. Two types of absorbers were used. A lead foil 0.0002 cm thick was placed along a diameter of the chamber. A lead collimator was used so that the source could see only the lead absorber. This arrangement was used to examine photoelectrons ejected by low energy gamma-rays. A second absorber used to study Compton recoils was essentially carbon. Paper 0.004 cm thick was smeared with carbon and dipped in paraffin. This was mounted in a manner similar to the lead. Recoil electrons in the forward direction only were examined.

Gamma-radiation was found associated with each of the iodine isotopes studied. Absorption measurements in lead have been carried out by Livingood and Seaborg.[6] Attempts to determine the energies of the gamma-rays associated with I^{126} and I^{128} were unsuccessful. With the strongest sources available and by using the largest solid angle possible, about one ejected electron was observed in fifty expansions.

Over 1000 photographs were taken of the electrons ejected from lead by the combined activities of I^{130} and I^{131}. With the strongest available sample, some 70 fair tracks were obtained. Indications of three lines were observed. The Compton recoils at 2580 $H\rho$ and the photoelectrons at 2900 $H\rho$ established quite definitely a gamma-ray having an energy of 0.59 Mev. Adding the energy of the scattered quantum to the recoil energy of 0.42 Mev corresponds to an energy of 0.58 Mev. Adding the K-electron binding energy of lead to the energy of the photoelectron yields 0.59 Mev. The second line occurred at about 1570 $H\rho$. From the relation of the Compton and photoelectric cross sections, it is seen that these electrons are probably entirely photoelectrons. Thus the energy of the gamma-ray is 0.27 Mev. A third line, rather uncertain, appeared at 0.46 Mev. This was identified by Compton recoils at 2100 $H\rho$ ($E=0.03$ Mev) and by photoelectrons at 2240 $H\rho$ ($E=0.36$ Mev). Applying the necessary corrections this indicates a gamma-ray energy of about 0.46 Mev.

At a later time additional pictures were taken when the I^{131} activity was present alone. Only a few tracks were obtained. However, these few tracks were grouped in two energy groups corresponding roughly to the 0.27- and 0.46-Mev lines. It was thus assumed that the line at 0.58 Mev was due to I^{130} and the other two lines due to I^{131}. The value of 0.58 Mev is in good agreement with the value of 0.6 Mev obtained by absorption measurements. The absorption value of 0.4 Mev for I^{131} means little for this case where there are two lines present.

The author is indebted to Professor J. M. Cork and other members of the nuclear physics group for their assistance. The chemical separations were performed by Mr. W. H. Sullivan. This work was made possible through a grant from the Horace H. Rackham Trust Fund.

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